Title
Heat losses in a CVD reactor for polysilicon production: comprehensive model and experimental validation.

Authors
A. Ramos*[1], A. Rodríguez[2], C. del Cañizo[1], J. Valdehita[1], J.C. Zamorano[1], A. Luque[1].

[1] Instituto de Energía Solar - Universidad Politécnica de Madrid, ETSI Telecomunicación, Avda. Complutense 30, 28040 Madrid, Spain
[2] Departamento de Ingeniería química, Fac. de Ciencias Químicas, Universidad Complutense, Avda. Complutense s/n, 28040 Madrid, Spain

***Corresponding author**
Alba Ramos

E-mail: alba.ramos@ies-def.upm.es

Telephone: (+34) 91 453 35 49
Fax: (+34) 91 544 63 41

Abstract

This work addresses heat losses in a CVD reactor for polysilicon production. Contributions to the energy consumption of the so-called Siemens process are evaluated, and a comprehensive model for heat loss is presented. A previously-developed model for radiative heat loss is combined with conductive heat loss theory and a new model for convective heat loss. Theoretical calculations are developed and theoretical energy consumption of the polysilicon deposition process is obtained. The model is validated by comparison with experimental results obtained using a laboratory-scale CVD reactor. Finally, the model is used to calculate heat consumption in a 36-rod industrial reactor; the energy consumption due to convective heat loss per kilogram of polysilicon produced is calculated to be 22-30 *kWh/kg* along a deposition process.

Keywords
B1. Polysilicon; A3. Chemical vapour deposition (CVD); A1. Heat transfer mechanisms; B2. Solar grade silicon

Nomenclature

P	Pressure (Pa)
T_s	Rod's surface temperature (K)
T_∞	Gas temperature far from the surface (K)
μ	Dynamic viscosity ($N \cdot s \cdot m^{-2}$)
β	Volumetric thermal expansion coefficient (K^{-1})
ν	Kinematic viscosity ($m^2 \cdot s^{-1}$)
ρ	Density ($kg \cdot m^{-3}$)
L	Characteristic length (m)
U_∞	Gas velocity far from the surface ($m \cdot s^{-1}$)
h	Convection coefficient ($W \cdot K^{-1} \cdot m^{-2}$)
c_p	Specific heat at constant pressure ($J \cdot kg^{-1} \cdot K^{-1}$)
k	Thermal conductivity ($W \cdot K^{-1} \cdot m^{-1}$)
J_α	Diffusion flux of species ($mol \cdot m^{-2} \cdot s^{-1}$)
H_α	Partial molar enthalpy of species ($J \cdot mol^{-1}$)
E	Energy per unit mass of gas ($J \cdot kg^{-1}$)
p	Static pressure (Pa)

g Gravity ($m \cdot s^{-2}$)

v Velocity ($m \cdot s^{-1}$)

t Time (s)

$\vec{\tau}$ Stress tensor (Pa)

r, θ, z Cylindrical coordinates (m, -, m)

1. Introduction

Chemical vapour deposition (CVD) processes are widely used for solid materials production from gaseous reactants. Several types of CVD processes exist and the resultant solid materials applications are varied: semiconductor devices, solar cells, optical fibres, coatings, etc. CVD processes are complex; thermodynamics, fluid dynamics, plasma physics, kinetics and chemistry are involved [1,2].

For silicon-based photovoltaic technology (PV), the chemical route through CVD, which is called "the Siemens process", presently dominates the production of purified Si (which is usually called polysilicon) [3,4]. The Siemens process consists of polysilicon deposition by a CVD process from trichlorosilane (TCS) and comprises two main steps. First, metallurgical silicon (MG Si) is transformed to TCS and purified, and, second, TCS is reduced into high purity silicon (the CVD process itself). This is a high energy consuming process; it is responsible for nearly one third of the final PV module cost [5,6,7]. Polysilicon production used to be monopolized by microelectronics, but, in the last decade, PV technology shifted microelectronics from the polysilicon market worldwide driven by its exponential growth [8]. An initial polysilicon shortage was followed in recent years by an oversupply scenario. In this context, the interest of polysilicon producers is focused on energy savings to make their way into the world market [9,10].

Among the multiple disciplines that comprise a CVD process, this paper is centered on energy consumption of the polysilicon CVD. To properly address heat loss of the polysilicon CVD process, deposition conditions, radiation heat emitted, electrical heating, convection and conduction heat losses and chemical reactions must be jointly studied. In industry, CVD of polysilicon is obtained through TCS decomposition conducted in a reactor chamber inside which a number of inverse U-shape polysilicon rods are heated by the Joule effect. Typically, rods are heated to around 1100*ºC* and the process pressure is 6 *bar*. The polysilicon CVD process consists of three steps: heating, polysilicon deposition and cooling down, the deposition step being the one accounting for almost all the energy consumption [11]. Previous published papers on CVD for polysilicon production are mainly centered on chemical reactions, setting aside heat transfer mechanisms [1,2]. On the contrary, this work is focused on the less studied field: heat transfer phenomena in CVD reactors.

In the following sections, the heat loss problem associated with polysilicon CVD is addressed; all contributions to the energy consumption are put forward and theoretical calculations are developed. A comprehensive theoretical model for heat losses is presented. A model for heat transfer, to evaluate energy losses due to different mechanisms, forms part of the comprehensive theoretical model. Theoretical calculations for heat conduction in gases and a novel model for convective heat loss are presented. In addition, a theoretical model for radiative heat loss -already published in [12]- is considered. Thereby, the theoretical energy consumption of the polysilicon deposition process is calculated. The present work adds to the previous work [12] by developing and validating a model for convective and conductive loss taking advantage of a CVD laboratory scale reactor.

2. Theoretical models for CVD heat loss phenomena in polysilicon production

To obtain the energy consumption of a CVD process, the energy balance equation is applied. It is obtained applying the law of conservation of energy to a volume element (VE) [13]. In the present case, a stationary volume element through which a fluid is moving can be defined. This volume element is a cylinder whose radius is defined by half the distance between either two seed rods; the center of the silicon rod is situated along the longitudinal axis of the cylinder and the reaction gas is flowing through the space between the rod's surface and the cylinder limits, Fig. 1.

The law of conservation of energy applied to the described system, per unit volume, leads to Eq. (1).

$$\frac{\partial}{\partial t}\left(\frac{1}{2}\rho v^2 + \rho E\right) + \nabla \cdot \left(\frac{1}{2}\rho v^2 + \rho E\right)\vec{v} = \nabla \cdot \left(k\nabla T - \sum_{\alpha=1}^{n} H_\alpha J_\alpha\right) - \nabla \cdot p\vec{v} - \nabla \cdot [\vec{\tau} \cdot \vec{v}] + \rho(\vec{v} \cdot \vec{g}) + S_c + S_r \quad (1)$$

The first term on the left side of Eq. (1) corresponds to the rate of variation of energy and the second one corresponds to the net rate of kinetic and internal energy transfer by convective transport. On the right side, the first terms, in between brackets, correspond to the net rate of heat transfer by molecular transport

due to temperature gradient (conduction) and enthalpy transport for an ideal gas, the second and third terms are the rate of work done on the system by molecular diffusion mechanisms (pressure and stress) and the forth by external forces. The term S_c corresponds to the heat of chemical reactions and S_r to the heat transfer by radiation [13].

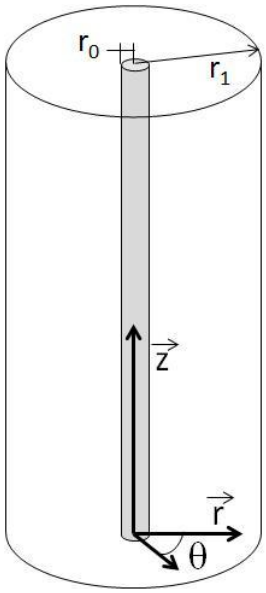

Fig. 1. VE for a single rod of a CVD process.

The heat of the reactions has a negligible influence on the gas temperature distribution. Therefore, the term S_c in Eq. (1) is hereafter disregarded. The reagent TCS is highly diluted, hydrogen absorbs changes and there is virtually no change in composition, therefore the diffusive source term $\sum_{\alpha=1}^{n} H_\alpha J_\alpha$ is also neglected. Polysilicon deposition is quite a slow process -it is in the range of microns per minute (*μm/min*)-; once the desired deposition temperature has been reached and after an equilibration time, the spatial distribution of temperatures does not change any further [14]. The problem is therefore considered to be a quasi-steady-state process, and the time derivative is zero. The kinetic terms, such as the external forces, can be neglected. The rate of heat generation by viscous dissipation is also negligible [14]. Heat transfer by radiation has a negligible influence on the gas temperature distribution, but the heat transfer by radiation that leaves one rod and reaches any other surface cannot be neglected. Thus, the term S_r corresponds to the heat exchanged by radiation between the different surfaces. Moreover, S_r is extracted from Eq. (1) and can be calculated separately evaluating the radiative heat flux exchanged between the rods and the reactor wall [12]. Finally, the thermal energy equation for molecular transport and mass transport mechanisms applied to the gas region of the VE is expressed in Eq. (2).

$$\nabla \cdot (\rho E)\vec{v} = \nabla \cdot (k \nabla T) \tag{2}$$

For the present problem, the transfer energy equation for molecular and convective transport is expressed in Eq. (3) in terms of temperature.

$$\nabla \cdot (c_p \rho\, T\vec{v}) - \nabla \cdot (k \nabla T) = 0 \tag{3}$$

It has to be noted that the electrical power consumed instantaneously along a CVD process must compensate all heat loss through the VE boundaries. From the reasoning above, the net heat loss is the sum of radiative, conduction and convection losses, where the latter two occur via gases.

The gas flowing along the rods' surface is responsible for conduction and convection heat loss, whereas the radiative heat flux between the different surfaces is accountable for radiation heat loss. Radiation heat loss is calculated separately from the thermal energy equation for molecular transport and molecular mechanisms, and is dealt with at the end of this section. Convection and conduction via gases must be jointly considered as shown in Eq. (3). In a gaseous system, heat transfer tends to be dominated by convection, and conduction through gases is neglected in many analyses. However, in the present case, both phenomena are studied together and neither can be disregarded, as will be shown below.

During polysilicon deposition the gas flows along the silicon rods. The rods' surfaces are kept at high temperature to allow the deposition reaction to occur, and the reactor wall is kept cooled at a constant temperature of around 100*ºC*. Therefore, the gas flowing through the reactor chamber presents a temperature distribution according to the rods and wall temperatures. The gas properties, such as thermal conductivity (k), dynamic viscosity (μ), specific heat at constant pressure (c_p) and density (ρ) are dependent on the gas temperature at the working conditions of pressure. In Eq. (3), all parameters can be known or expressed as functions of temperature. Therefore, the temperature distribution can be obtained. Once the temperature distribution is known, the convection and conduction heat loss can be calculated.

First, Eq. (3) is expressed in cylindrical coordinates. A temperature gradient exists in the radial direction (r). Due to the cylindrical symmetry of the problem, and disregarding inlet and outlet effects, $T(\theta)$ and $T(z)$ are assumed to be constant. The gas properties (k,μ,c_p,ρ) are calculated using the equations for the gas mixture properties appearing in [15]. When performing the calculations presented in this work, it has been checked that all values obtained are in agreement with other bibliographic sources [15,16]. It must be pointed out that the gas's properties vary appreciably with temperature variations, but not with pressure variations between 1 and 6 *bar*. The gas velocity is assumed to move parallel to the rod's surface ($\vec{v} = U_\infty \vec{z}$). U_∞ is considered to be constant and equal to the free stream velocity. Gas velocity at the rods' surface and the reactor wall is zero to accomplish continuity and momentum equations at that point. Nevertheless, the boundary layer thickness is at least two orders of magnitude lower than the VE radius (r_1); its effect on heat loss by convection will be considered in the model, but, for simplicity, it can be neglected for this first calculation to obtain the gas temperature distribution.

By developing Eq. (3) and considering the particular conditions of the problem, Eq. (4) is obtained. r corresponds to the radial coordinate and the terms k', c_P' and ρ' correspond to $\partial k/\partial T$, $\partial c_P/\partial T$, and $\partial \rho/\partial T$, respectively. The boundary conditions needed to solve this differential equation are the temperature at the rod surface, $T(r = r_0) = T_0$, and at the boundaries of the volume element (VE), $T(r = r_1) = T_1$.

$$k\frac{\partial^2 T}{\partial r^2} + k'\left(\frac{\partial T}{\partial r}\right) - r\,U_\infty \frac{\partial T}{\partial r}(\rho\, c_P + c_P' T\rho + c_P T\rho') = 0 \tag{4}$$

The functions $\rho(r)$, $k(r)$ and $c_P(r)$ are known and their derivatives are three orders of magnitude lower than their respective absolute values: $k' = (O)^3 k$, $c_P' = (O)^3 c_P$ and $\rho' = (O)^3 \rho$. Therefore, Eq. (4) is simplified leading to Eq. (5).

$$k\frac{\partial^2 T}{\partial r^2} + -\frac{\partial T}{\partial r} c_P r\, U_\infty \rho = 0 \tag{5}$$

2.1 Conductive heat loss

Once the temperature distribution in the gas surrounding a silicon rod is obtained, conductive heat loss through the gases can be calculated from the heat conduction theory if the geometry, boundary conditions and gas properties are known [17]. The process of heat transfer by conduction is governed by partial differential equations, which can be solved through standard methods [18]. Eq. (6) shows the general expression for conductive heat loss in watts per unit area at steady state without internal heat generation. Again the boundary conditions needed to solve this equation are the temperature at the rod surface, $T(r = r_0) = T_0$, and at the VE boundaries, $T(r = r_1) = T_1$.

$$Q_{conduction} = -k\nabla T \tag{6}$$

Due to the cylindrical symmetry of the problem, Eq. (6) can be reduced for the present case to Eq. (7).

$$Q_{conduction} = -\frac{k}{r}\frac{\partial}{\partial r}(rT) \tag{7}$$

2.2 Convective heat loss

Convective heat loss can be calculated starting from the gas's temperature distribution. This heat transferred between a surface and a moving fluid is a combination of molecular diffusion due to the concentration gradient (perpendicular to the gas flow) and bulk motion of molecules (parallel to the gas flow). The flow velocity near the surface is low (diffusion dominates), and as we move away of the surface the bulk motion increases and it becomes the dominating force. As described above, the velocity of the fluid layer in contact with the surface is zero (non slip condition); then, the heat transferred by convection and conduction from the heated surface to the fluid is defined as the heat transferred by conduction inside the solid that reaches the solid's surface. To calculate this heat transferred by conduction, the thermal

conductivity of the solid surface and the thermal boundary layer (BL) need to be known. Thermal BL calculations are not straightforward [19,20]. Heat transfer loss by convection, in watts per unit area, can be calculated if the convection coefficient (h), the temperature at the rod's surface (T_s) and the average temperature of the free stream (T_∞) are known:

$$Q_{convection} = h \cdot (T_s - T_\infty) \tag{8}$$

The purpose of the model is to find or approximate h for the geometry and flow conditions under investigation in order to apply Eq. (8). Different procedures exist which purse the empirical constant h: a combination of dimensional analysis and experiments, exact mathematical solutions of the boundary layer equations, approximated analysis of the boundary layer equations (integral methods), heat transfer and linear momentum analogy and numerical analysis [21]. Exact solutions only exist for a few geometries and flow conditions; according to the literature exact solutions for ours conditions exit. A known analytical solution supported with experimental data has been found as valid for modeling the convection phenomenon in a CVD reactor.

In the following subsection, a model is presented for the calculation of convective heat loss, which is based on a known analytical solution: cylinder in parallel flow. To the authors' knowledge this is the first time such a model has been applied to convective heat loss in a CVD reactor. First, flow conditions must be analyzed. Flow conditions are defined by means of several dimensionless numbers. The numbers relevant to the present case are Reynolds (Re), Prandtl (Pr), Grashof (Gr) and Nusselt (Nu). These are defined in Eqs. (9-12):

$$Re = \frac{U_\infty L}{\nu} \tag{9}$$

$$Pr = \frac{C_p \mu}{k} \tag{10}$$

$$Gr = \frac{g\beta(T_s - T_\infty)\, L^3}{\nu^2} \tag{11}$$

$$Nu = \frac{h \cdot L}{k} \tag{12}$$

For given flow conditions, Re gives a measure of the ratio of inertial forces to viscous forces, Pr gives the ratio of momentum diffusivity (kinematic viscosity) to thermal diffusivity, Gr approximates the ratio of the buoyancy to viscous force acting on a fluid and Nu is the ratio of convective to conductive heat transfer normal to the surface-fluid interface. Inlet gas conditions define the free stream velocity (U_∞) and the volumetric thermal expansion coefficient ($\beta = 1/T_\infty$). The geometry of the problem, gas properties and boundary conditions -pressure (P) and temperatures (T_s,T_∞)- must be known.

Convective heat transfer may take the form of either forced or natural (free) convection. Forced convection occurs when a fluid flow is induced by an external force and natural convection is caused by buoyancy forces due to density differences caused by temperature variations in the fluid. From Navier Stokes' boundary-layer equation, it is realized that the relationship between Gr and Re shows a qualitative indication of the influence of buoyancy on the forced convection flow conditions. Exact solutions only exist if either of the following three conditions are fulfilled [20,22]:

$$a)\ Gr \leq 0.150 Re^2 \ \ and \quad Gr < 10^8$$

$$b)\ Gr > 0.007 Re^{2.5} \ \ and \quad Gr > 10^8$$

$$c)\ Gr < 0.0016 Re^{2.5} \ \ and \quad Gr > 10^8$$

Cases a) and c) are considered forced convection (natural convection below 10%). Case b) corresponds to natural convection (forced convection below 10%). Relations in between cases b) and c) are considered combined convection; no exact solutions exist for this combined case. Due to the nature of the CVD process, both forced and natural convection phenomena must be considered [20,22]. The flow conditions determine whether or not an exact solution, such that $Nu = f(Re, Gr, Pr \ldots)$, can be found.

It is necessary to distinguish laminar from turbulent flow in natural or forced convection regimens. For $Re > 10^6$ and $Gr > 10^9$, it is assumed that there is turbulent flow, and for $Re < 10^5$ and $Gr < 5 \cdot 10^8$, there is laminar flow [19]. Thus, depending on the flow regime, and for a fixed geometrical conditions, once the analytical solution is known, the relation between the Nu, Pr, Gr and Re numbers is also known. The last three dimensionless numbers can be calculated from the geometry of the problem, the P and T conditions and the gas properties. $Nu = f(Pr, Re)$, $Nu = f(Pr, Gr)$ or $Nu = f(Pr, Re, Gr)$ depend on the flow

conditions: forced, natural or combined forced and natural convection, respectively. Thus, Nu can be calculated. h can be calculated from Eq. (12), given that the characteristic length (L) and the thermal conductivity of the gas (k) are known. Notice that accuracy of h calculations tends to be under 20% due to the experimental data variation [19].

2.3 Radiative heat loss

A theoretical model for radiative heat loss calculations has been developed at our institute, described in detail and applied to different reactor configurations in [12,23] and validated with the help of a CVD laboratory prototype in [11]. In those works, the radiative model demonstrated that the overall power consumption shows a string dependence on the rods' surface temperature. It was recommended that power consumption can be reduced by decreasing the reactor wall emissivities and increasing the geometrical design compactness. The same model has been used for the calculation of radiative heat loss in the present work.

3. Validation of the convection heat loss model on a laboratory prototype

Complexity and high energy consumption characterize the CVD process for polysilicon production. Research on the polysilicon CVD process is not easily affordable at the industrial scale due to the large amount of time and money required. However, a laboratory scale reactor capable of reproducing an industrial reactor's working conditions allows this research to be performed in a more accessible way. At the *Instituto de Energía Solar (IES)*, there is a laboratory prototype of the Siemens type. Many rods can be positioned within the prototype's chamber, and there are gas inlets and outlets and the bottom and top respectively. In this work, experiments have been conducted in the CVD laboratory using a single rod configuration, which is shown schematically in Fig. 2. Industrial working conditions can be reproduced with the prototype (except the working pressure); thus, prototype results can be satisfactorily extrapolated to the industry [24]. It should be observed that a single rod configuration has a lower compactness (i.e. a larger distance between the rod surface and the reactor wall) than the many-rod configurations favored in industry. The effect of this on the total power consumption is also discussed in this and the following sections.

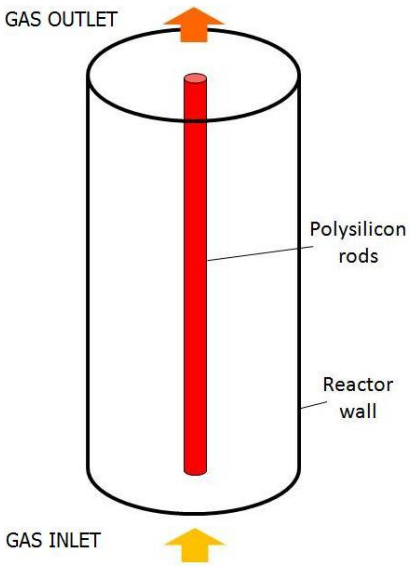

Fig. 2. Schematic of a single rod configuration for the CVD laboratory prototype.

3.1. CVD laboratory prototype convection heat loss

The case of one single rod placed inside the reactor chamber is first analyzed. All the parameters needed to calculate Re, Pr and Gr are known: the inlet gas velocity, the temperatures, the gas properties, etc. The inlet gas velocity is assumed constant along the deposition process, the gas flow is kept constant and the flow area variation along the process can be neglected. The values obtained for these dimensionless numbers for different gas temperatures are shown in Table 1. The mass ratio, H_2:TCS, of the reactive gases for the values below is 1:1.34, and the working pressure is 1 *bar*.

Table 1. Pr, Re and Gr for the CVD laboratory prototype at different temperatures.

T [ºC]	Pr [-]	Re [-]	Gr [-]
50	0.397	109.4	6.53E+09
75	0.397	96.2	3.29E+09
100	0.396	85.4	1.89E+09
125	0.396	76.4	1.18E+09
150	0.395	68.8	7.78E+08
175	0.394	62.3	5.33E+08
200	0.393	56.7	3.76E+08
225	0.392	51.9	2.73E+08
250	0.391	47.7	2.01E+08
275	0.390	44.1	1.51E+08
300	0.389	40.8	1.15E+08

The relationship $Gr > 0.007 \cdot Re^{2.5}$ is satisfied for all process conditions along a deposition process. Therefore, based on [20,22], the flow conditions of the CVD process in the laboratory prototype are natural convection. Being that $Gr > 10^9$ for gas temperatures below 125ºC and $Gr < 5 \cdot 10^8$ for gas temperatures above 175ºC, and knowing from experimental experience that the free-stream temperature is around 225ºC, then there is laminar flow for the process conditions. In light of this, and given the cylindrical symmetry of the CVD prototype, we may employ the empirical expression for $Nu(Gr, Pr)$ presented in [20]:

$$Nu = 0.68 \cdot Pr^{0.5} \cdot \left(\frac{Gr}{(0.925 + \mathrm{Pr})}\right)^{0.25} \tag{13}$$

The convection coefficient (h) is obtained from Eqs. (12) and (13). In Table 2, the values obtained for Nu and h are shown. Notice that h varies depending on the z coordinate; the values presented are averaged along the rod's length.

Table 2. Nu and h values for the CVD laboratory prototype at different temperatures.

T [ºC]	Nu [-]	h [$W / K{\cdot}m^2$]
175	60.5	26.1
200	55.4	24.9
225	51.1	23.9
250	47.3	23.0
275	44.0	22.2
300	41.1	21.5

For the process conditions of the CVD prototype, the convection coefficient is such that $21 < h < 38\, W/Km^2$. Heat loss due to convection can be calculated by means of Eq. (8). Notice that the convection coefficient varies with time since the process conditions vary; thereby, the heat loss due to convection will vary along a deposition process.

3.2 Comparison of theoretical and experimental results

In this subsection, theoretical and experimental results for three deposition processes are compared, with the aim of validating the model. Three different experiments were conducted with the CVD prototype in the single rod configuration with rod surface temperatures of 1000ºC, 1100ºC and 1150ºC (experiments 1, 2 and 3, respectively). The above-described model for total heat loss was also employed under the same set of conditions. Results shown below refer to the polysilicon deposition step, the largest energy consumer.

In the experiments, the power consumption is measured throughout the process. The temperature of the rod (T_o) and the wall (T_1) and the temperature of the gases inside the reactor chamber -among others- are monitored. Along the deposition process, certain parameters vary as a consequence of the increase of the

radiation area; thus, the total power consumed increases with time. The power supplied changes in short steps and not steadily because of the control algorithm causing oscillations of the rods' temperature; consequently, there are oscillations in the measured power consumption. Temperature oscillations are $\pm 15^{o}C$, resulting in a maximum experimental error of the power consumption measured of 4.5%. The initial diameter of the seed rods is between 0.73 and 0.76 *cm* and the length is 53 *cm*. Depending on the rods' surface temperature (for a fixed TCS and H_2 mass ratio) the deposition rate of polysilicon will vary. It is known that higher surface temperatures lead to higher deposition rates, thus shortening the deposition processes for the same final volume of silicon obtained at the expense of higher energy consumption [25]. The duration of the CVD processes conducted at the IES laboratories is between 5 and 10 hours and the growth rates are in the range of 2.1-3.9 $\mu m/min$. These growth rates measured experimentally are 4-5 times lower than the typical numbers in industry due to the low working pressure of the prototype and the bad compactness of the rods rearrangement. Further details of the experimental conditions can be found in [23,25].

At the laboratory, the rod diameter growth cannot be measured during a deposition process but it can be calculated when the process ends. For the theoretical calculations presented below, equations presented in section 2 are solved for a number of diameters, starting with the initial rod diameter and ending with the final one.

The total power consumption calculated theoretically and the experimental data measured at the laboratory are compared in the figures below. Figs. 3, 4 and 5 correspond to the experiments 1, 2 and 3, respectively.

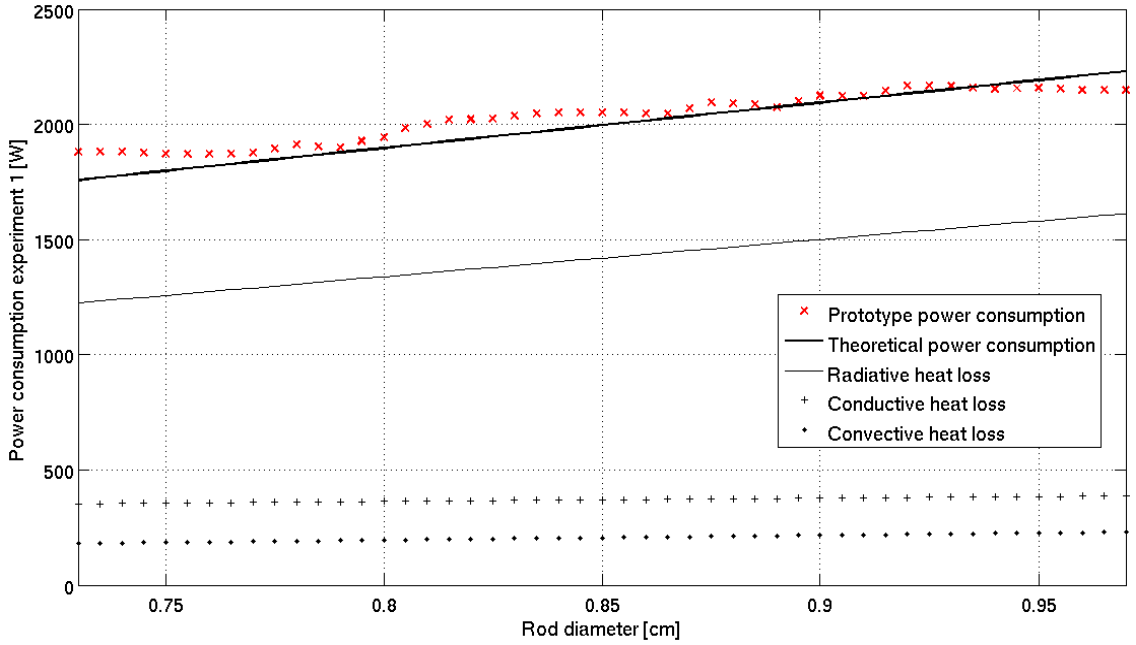

Fig. 3. Experimental (crosses) and theoretical (solid line) power consumption. Experiment 1: rod's surface at $1000^{o}C$.

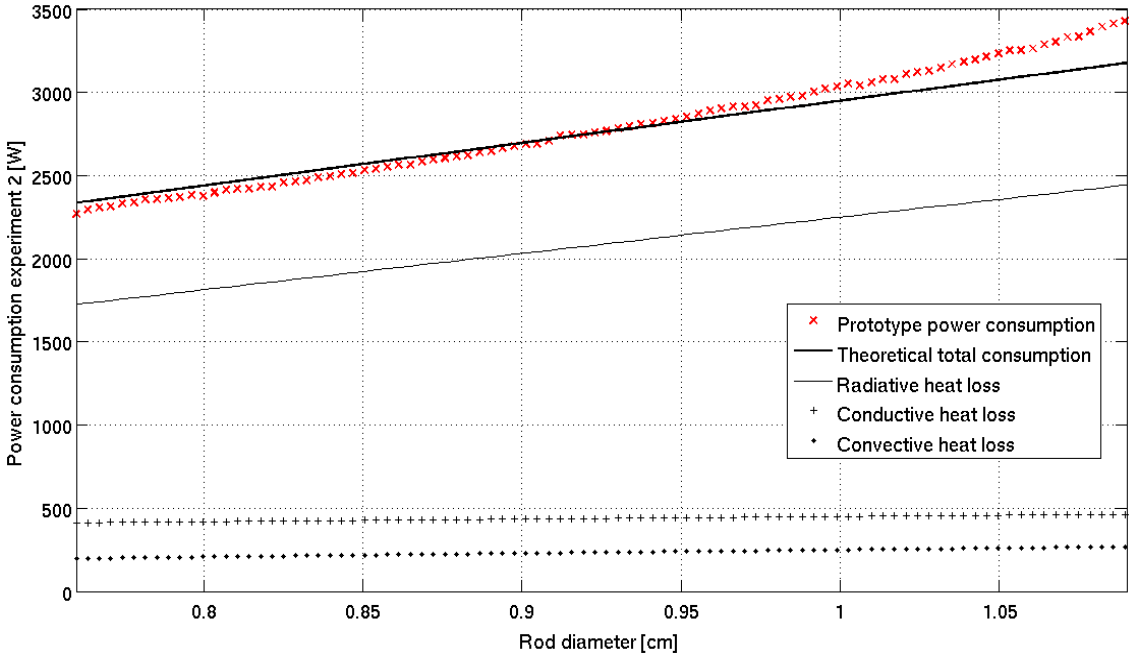

Fig. 4. Experimental (crosses) and theoretical (solid line) power consumption. Experiment 2: rod's surface at 1100^{o}C.

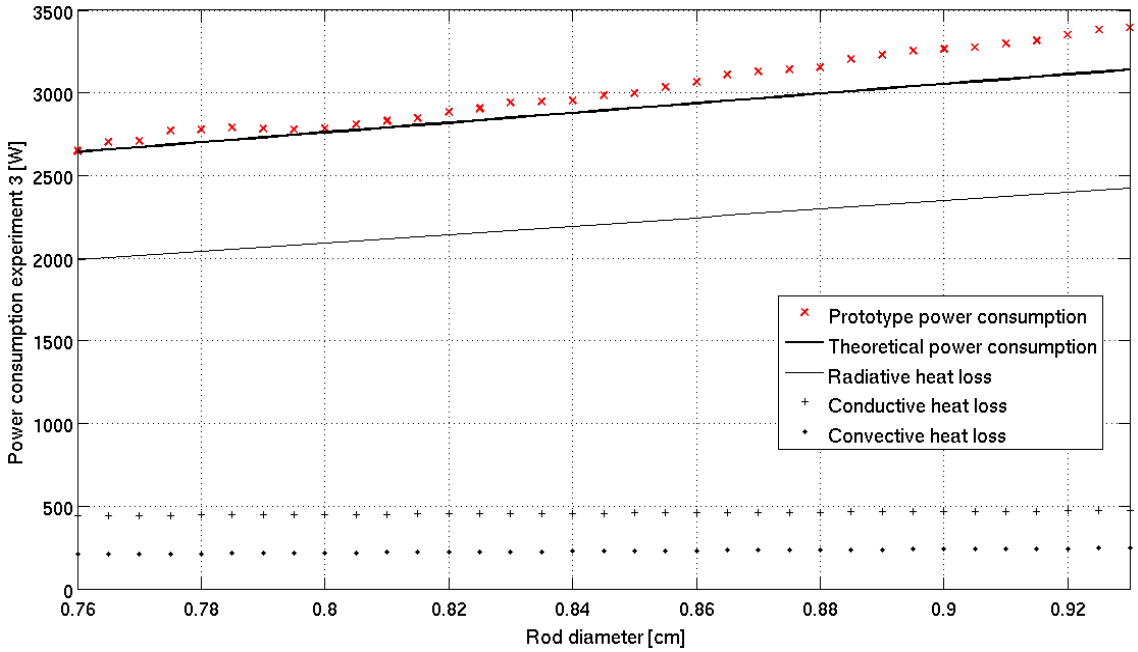

Fig. 5. Experimental (crosses) and theoretical (solid line) power consumption. Experiment 3: rod's surface at 1150^{o}C.

From the Figs. 3, 4 and 5, good agreement is observed between the measured and predicted values. The maximum difference between the experimental measurements and theoretical calculations is 4%, 2.5% and 1% in experiments 1, 2 and 3, respectively, which are within the experimental error. The average of the total power consumption measured, the predicted one by the theoretical model and the difference between both values are presented in Table 3. The difference between the values averaged over the deposition process is under 1.7%, 1.4% and 4% in experiments 1, 2 and 3, respectively. Radiation heat loss is responsible for 63.6-70.3% of the total energy consumption and conductive and convective phenomena are responsible for 13.7-16.6% and 7.2-9.3%, respectively. These percentages are in agreement with those published in [11]. In addition, conductive and convective heat losses are of the same order of magnitude, therefore, it is confirmed that both phenomena should be taken into account for heat loss calculations as stated in section 2.

Table 3. Measured and predicted total power consumption.

Experiment	Power measured [*W*]	Power predicted [*W*]	Difference [%]
1	2031	1996	1.7
2	2805	2767	1.4
3	3013	2892	4.0

It has to be noted that the absolute numbers as kilowatts-hour per kilogram of silicon (*kWh/kg*) obtained with the laboratory prototype are in the range of 350-400 *kWh/kg*, several times higher than those at the industry. The bad compactness of the single-rod configuration and the process pressure, which is 6 times lower than in industry [3], explains this high energy consumption. Also, the relation between radiation heat loss and convection heat loss for the prototype is in the range of 7.5-8.8, to be compared to the ratio for industry, which is presented in the next section.

4. Extension to industrial scale CVD reactor

Nowadays, overall energy consumption of the CVD process itself for large capacity polysilicon plants is said to be in the range of 45-50 kWh/kg. There are no detailed data of radiation, convection, and neither conduction heat losses coming from industry; therefore, the purpose of this section is to show an example of convection heat loss of a 36-rods industrial CVD reactor according to the model presented in this work.

The geometry and process parameters of the 36-rod industrial reactor are known. In contrast with the prototype, inlet gas conditions vary along the deposition process. The range of values obtained for Re, Pr and Gr is shown in Table 4. The molar ratio of the reactive gases for the values below is 14% mol TCS and 86% mol H_2 and the working pressure is 6 *bar*.

Table 4. Re, Pr and Gr for a 36-rod CVD industrial reactor at different temperatures.

T [^oC]	Re $_{max}$ [-]	Re $_{min}$ [-]	Pr [-]	Gr [-]
200	6786.2	437.2	0.208	4.15E+12
225	6508.8	419.3	0.209	2.98E+12
250	6256.2	403.1	0.209	2.18E+12
275	6025.6	388.2	0.209	1.63E+12
300	5813.7	374.6	0.210	1.23E+12
325	5618.2	362.0	0.210	9.44E+11
350	5437.3	350.3	0.210	7.32E+11
375	5269.6	339.5	0.210	5.73E+11
400	5113.4	329.4	0.211	4.53E+11
425	4967.5	320.0	0.211	3.61E+11
450	4831.0	311.3	0.211	2.89E+11
475	4702.8	303.0	0.211	2.33E+11
500	4582.4	295.2	0.212	1.89E+11
525	4468.8	287.9	0.212	1.54E+11
550	4361.5	281.0	0.212	1.26E+11

For the industrial CVD reactor, the conditions $Gr > 0.007 \cdot Re^{2.5}$ and $Gr > 10^9$ are fulfilled at all temperatures. Therefore, the flow conditions of an industrial CVD reactor are natural convection and turbulent flow for the process conditions shown. For these conditions, the empirical expression for $Nu(Gr, Pr)$ presented in [20] is:

$$Nu = 0.13 \cdot (Gr \cdot Pr)^{0.33} \tag{14}$$

The convection coefficient (h) is obtained from Eqs. (12) and (14). For the process conditions in the industrial CVD reactor, the convection coefficient is such that $58 < h < 85\,W/Km^2$. Nu and h values are presented in Table 5.

Table 5. Nu and h values for a 36-rod CVD industrial reactor at different temperatures.

T [^oC]	Nu [-]	h [W / $K{\cdot}m^2$]
200	1130.4	123.7
225	1013.6	115.9
250	915.1	109.1
275	831.0	103.1
300	758.3	97.8
325	694.9	92.9
350	639.2	88.5
375	589.9	84.4
400	546.0	80.7
425	506.6	77.2
450	471.1	74.0
475	439.0	70.9
500	409.8	68.1
525	383.2	65.4
550	358.7	62.8

The convection coefficient varies with time as the process conditions do; therefore, the heat loss due to convection will vary in consequence along a deposition process. Heat loss due to convection is calculated by means of Eq. (8).

The model for convection heat loss can be applied to any type of industrial reactor by reproducing the preceding steps. The model is therefore an important tool for convective heat loss calculations and, ultimately, for the total energy consumption of the CVD process.

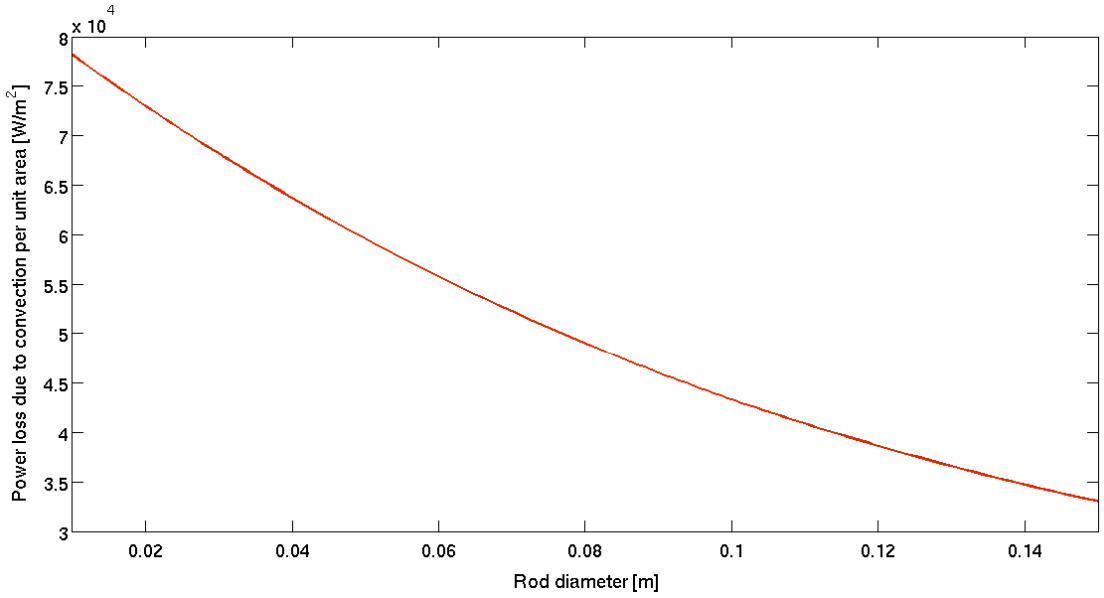

Fig. 6. Power loss due to convection per unit area for a 36-rods industrial reactor.

As an example, the model has been applied assuming a polysilicon deposition rate of 12 *μm/min*, a homogenous rod temperature of 1100^oC and a range of temperatures of the free stream between 400-525 ^oC. The calculated power consumption due to convection heat loss along a deposition process is in the

range of 22-33 *kWh/kg*. In Fig. 6, the power loss due to convection per unit area for a 36-rod industrial reactor along a deposition process is presented. In Fig. 7, the power loss due to convection per rod along a deposition process is also presented. Notice that temperature of the main stream increases as the diameter of the rods do, therefore the heat loss due to convection per unit area decreases (Eq. (8)) justifying the curve's shape in both figures.

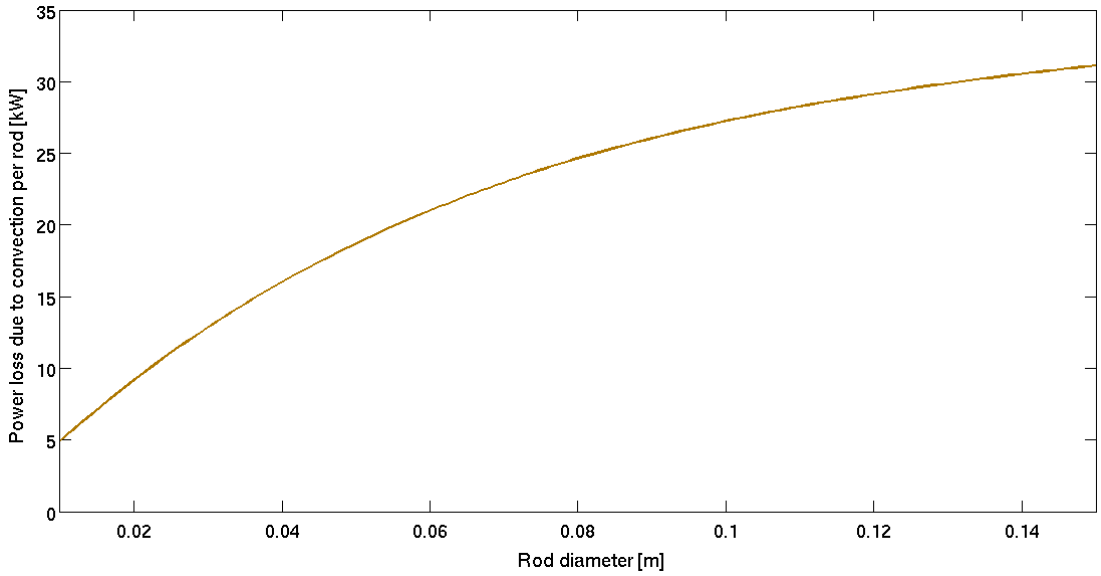

Fig. 7. Power loss due to convection per rod along a deposition process for a 36-rods industrial reactor.

For the same reactor under the same conditions, the radiative loss was calculated in [11] to be 25 *kWh/kg*. From these results, the relation between radiative and conductive heat losses is in the range of 0.88-1.20. The improvement in compactness in order to decrease radiation heat loss is evidenced.

Despite the difficulty to find truly and detailed numbers of power consumption in industry processes, theoretical calculations for radiation and convection heat losses for an industrial reactor presented in this paper are in agreement with the lumped figures reported by industry.

Conductive heat loss for a 36-rods reactor is calculated applying the heat conduction theory [17]. Exact calculations should consider the whole reactor and may need CFD software for its resolution; therefore those calculations are out of the scope of this work.

5. Conclusions

In this paper, heat consumption modeling in a CVD reactor for polysilicon production is addressed. This work focuses on the heat loss in the CVD process. The total energy conservation equation is applied to the problem and the phenomena responsible for heat loss are identified, namely, radiation and conduction and convection via gases.

Theoretical models for radiation, gases conduction and convection heat loss calculations are presented. The theoretical model for radiation heat loss presented was already validated and gases conduction heat loss is calculated by means of the heat conduction theory. A novel theoretical model for convection heat loss calculations in a CVD reactor has been developed in this work.

Taking advantage of a CVD laboratory prototype reactor, a number of experiments are conducted and the total power consumption of the polysilicon deposition process is monitored. Moreover, theoretical models for radiation and gases conduction and convection heat loss are applied to three experiments particular cases.

Comparing experimental and theoretical results, the model for gases convection heat loss is validated. In addition, radiation phenomenon arises as the main responsible of the total heat loss followed by gases conduction and convection phenomena, respectively; none of the latter two can be disregarded.

The heat-consumption theoretical models presented in this work can be applied not only to laboratory scale CVD reactors, but for industrial scale ones. Hence they are an important tool for research on heat loss in the polysilicon CVD process

Acknowledgement

The Spanish Ministerio de Economía y Competitividad is greatly acknowledged for its support through project IPT 2012-0340-120000.

Alexander Mellor is acknowledged for his contribution to the manuscript revision.

References

[1] Chris R. Kleijn, Computational modeling of transport phenomena and detailed chemistry in chemical vapor deposition – a benchmark solution, Thin Solid Films 365 (2000), 294-306.

[2] Z. Pan, Effect of Boundary Layers on Polycrystalline Silicon Chemical Vapor Deposition in a Trichlorosilane and Hydrogen System, Chinese Journal of Chemical Engineering,19(1), 2011.

[3] S. K. Chunduri, Innovations in inertia: Market survey on Siemens-type CVD reactors, Photon International, 4 (2013), 114-126.

[4] C. Wang, T. Wang, P. Li, Z. Wang, Recycling of $SiCl_4$ in the manufacture of granular polysilicon in a fluidized bed reactor, Chemical Engineering Journal 220 (2013).

[5] B. Ceccaroli, O. Lohne, Solar grade silicon feedstock, in: Handbook of Photovoltaic Science and Engineering, 2nd Ed., 2011, pp. 169–217.

[6] E. Alsema, M. de Wild-scholten, Reduction of the environmental impacts in crystalline silicon module manufacturing, in: 22nd European Photovoltaic Solar Energy Conference and Exhibition, 2007, pp. 829–836.

[7] S. Singer, Photovoltaics: Getting Cheaper, Eco Queen of Green, 2007.

[8] K. Hesse, E. Schindlbeck, E. Dornberger, M. Fischer, Status and development of solar-grade silicon feedstock, in: 24th European Photovoltaic Solar Energy Conference, 2009, pp. 883–885.

[9] M. Meyers, Silicon 2012: the new Si game, in: 10th Solar Silicon Conference, 2012.

[10] No End in Sight for Polysilicon Oversupply, Technical Report, NPD Solarbuzz (2012).

[11] A. Ramos, C. del Cañizo, J. Valdehita, J.C. Zamorano, A. Luque, Radiation heat savings in polysilicon production: Validation of results through a CVD laboratory prototype, Journal of Crystal Growth 374 (2013) 5-10.

[12] G. del Coso, C. Cañizo, A. Luque, Radiative energy loss in a polysilicon CVD reactor, Solar Energy Materials & Solar Cells 95 (2011), 1042–1049.

[13] R. Bird et al., Transport phenomena (2nd edition), John Wiley & Sons, 2007.

[14] D.E. Rosner, Transport Processes in Chemically Reacting Flow Systems, Dover publications, New York, 2000.

[15] B. E. Poling, J. M. Prausnitz, J. P. O'Connell, Properties of gases and liquids (5th edition), McGraw-Hill Education, 2001.

[16] http://www.fiz-chemie.de/infotherm/servlet/infothermSearch (last accessed: 20th November 2013).

[17] F. Kreith, M. Bohn, Principles of heat transfer, Thomson 6th ed., 2002, pp. 73–95.

[18] H. S. Carslaw, J. c. Jaeger, Conduction of Heat in Solids, 2nd ed., Oxford University Press, London, 1986.

[19] A.J. Chapman, Heat transfer, 4th ed., Macmillan, New York, 1984.

[20] H. Schilichting, Boundary-Layer theory, 6th ed., Mc Graw Hill series in mechanical engineering, 1968.

[21] F. Kreith, M. Bohn, Principles of heat transfer, Thomson 6th ed., 2002, pp. 233–342.

[22] B. Gebhart, Heat transfer, 2nd Ed., chap. 8, McGraw-Hill, New York, 1970.

[23] A. Ramos, C. del Cañizo, J. Valdehita, J.C. Zamorano, A. Rodríguez, A. Luque, On track for Solar Grade Silicon through a Siemens process-type laboratory reactor: operating conditions and energy savings, in: Silicon for the Chemical and Solar Industry XI Conference, 2012.

[24] G. del Coso, Chemical Decomposition of Silanes for the Production of Solar Grade Silicon, Ph.D. Thesis, Universidad Politécnica de Madrid - ETSIT, 2010.

[25] A. Ramos, C. del Cañizo, J. Valdehita, J.C. Zamorano, A. Rodríguez and A. Luque, Exploring polysilicon deposition conditions through a laboratory CVD prototype, Physica Status Solidi C 9 (2012), 10-11.